

**PESTICIDAL COMPOSITIONS CONTAINING
PLANT ESSENTIAL OILS AGAINST BEETLES**

CROSS REFERENCE TO RELATED APPLICATION

This application claims the benefit of Provisional Application Serial No. 60/147,421, which was filed August 6, 1999.

FIELD OF THE INVENTION

The present invention relates, in general, to pesticidal compositions containing plant essential oils and/or derivatives thereof against beetles. In one aspect, the present invention relates to pesticidal compositions containing one or more plant essential oils and/or derivatives thereof to be used as a contact pesticide in containers or cartons where food and other products are stored. In another aspect, the present invention relates to pesticidal compositions containing one or more plant essential oils and/or derivatives thereof to be used as a fumigant pesticide in containers or cartons where food and other products are stored. In a further aspect, the present invention relates to a method for controlling stored product pests by the application of pesticidally effective amounts of the pesticidal compositions to the container or carton in which food and other products are stored.

BACKGROUND OF THE INVENTION

Pests (invertebrates, insects, arachnids, larvae thereof, etc.) are annoying to humans for a myriad of reasons. They have annually cost humans billions of dollars in crop losses and in the expense of keeping them under control. For example, the losses caused by pests in agricultural environments include decreased crop yield, reduced crop quality, and increased harvesting costs.

Over the years, synthetic chemical pesticides have provided an effective means of pest control. For example, one approach teaches the use of complex, organic insecticides, such as disclosed in U.S. Pat. Nos. 4,376,784 and 4,308,279. Other

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approaches employ absorbent organic polymers for widespread dehydration of the insects. See, U.S. Pat. Nos. 4,985,251; 4,983,390; 4,818,534; and 4,983,389. Use of inorganic salts as components of pesticides has also been tried, as disclosed in U.S. Pat. Nos. 2,423,284 and 4,948,013, European Patent Application No. 462 347, Chemical Abstracts 119(5):43357q (1993) and Farm Chemicals Handbook, page c102 (1987).

However, it has become increasingly apparent that the widespread use of synthetic chemical pesticides has caused detrimental environmental effects that are harmful to humans and other animals. For instance, the public has become concerned about the amount of residual chemicals that persist in food, ground water and the environment, and that are toxic, carcinogenic or otherwise incompatible to humans, domestic animals and/or fish. Moreover, some target pests have even shown an ability to develop immunity to many commonly used synthetic chemical pesticides. In recent times, regulatory guidelines have encouraged a search for potentially less dangerous pesticidal compositions via stringent restrictions on the use of certain synthetic pesticides. As a result, elimination of effective pesticides from the market has limited economical and effective options for controlling pests. As an alternative, botanical pesticides are of great interest because they are natural pesticides, i.e., toxicants derived from plants that are safe to humans and the environment.

With respect to protecting stored products, including food, from insects, this safety issue is even more important. Some of the major stored food products affected by beetles include, but are not limited to, flour, grain, wheat, barley, corn, pasta, cereal, pet food, and others. Since the pesticides will come in contact with the stored food products to some extent, it is essential that the pesticides be safe for mammals and do not persist for very long, but rather break down very easily. Stored food products have long been subject to infestation by beetles, e.g., red flour beetle, confused beetle, maize weevil, sawtoothed grain beetle, drugstore beetle, etc., that contaminate foodstuffs, thereby making them unfit for consumption and/or causing certain adverse reactions in

Accordingly, there is a great need for novel pesticidal compositions, containing no pyrethrum, synthetic pyrethroids, chlorinated hydrocarbons, organo phosphates, carbamates and the like, that can be effectively used inside the containers or cartons for stored food products as both a contact and fumigant pesticide against beetles. In addition, there is a need for a method of treating the containers or cartons to kill and repel beetles, thereby preserving the stored food products contained therein.

A primary object of the present invention is to provide novel pesticidal compositions for containers or cartons in which food and other products are stored.

A further object of the present invention is to provide novel pesticidal compositions that contain one or more plant essential oils and/or derivatives thereof, natural or synthetic, as a fumigant pesticide in containers or cartons against beetles.

It is also an object of the present invention to provide a pesticidal composition and method for mechanically and neurally controlling beetles.

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It is still another object to provide a pesticidal composition and method that has a pleasant scent or is unscented, and that can be applied without burdensome safety precautions.

It is still another object to provide a pesticidal composition and method as described above which can be inexpensively produced or employed.

It is yet another object of the invention to provide a pesticidal composition and method to which pests cannot build immunity.

The above and other objects are accomplished by the present invention, which is directed to pesticidal compositions comprising plant essential oils and/or derivatives thereof, natural or synthetic, in admixture with suitable carriers. In addition, the present invention is directed to a method for controlling beetles by applying a pesticidally effective amount of the above pesticidal compositions to the container or carton in which food and other products are stored.

Additional objects and attendant advantages of the present invention will be set forth, in part, in the description that follows, or may be learned from practicing or using the present invention. The objects and advantages may be realized and attained by means of the instrumentalities and combinations particularly recited in the appended claims. It is to be understood that the foregoing general description and the following detailed description are exemplary and explanatory only and are not to be viewed as being restrictive of the invention, as claimed.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

All patents, patent applications and literatures cited in this description are incorporated herein by reference in their entirety. In the case of inconsistencies, the present disclosure, including definitions, will prevail.

In one embodiment, the present invention provides a pesticidal composition comprising, in admixture with a suitable carrier and optionally with a suitable surface active agent, comprising one or more plant essential oil compounds and derivatives

BOOK REVIEW

BOOK REVIEW

It will be appreciated by the skilled artisan that the pesticidal compositions of the present invention unexpectedly exhibit excellent pesticidal activities using one or more U.S. F.D.A. approved plant essential oils, in lieu of conventional pesticides which are not safe for use in food containers or cartons. Without wishing to be bound by the following theories, it is believed that plant essential oils antagonize a pest's nerve

Use of pesticidal compositions of the present invention generally results in 90-100% mortality on contact, and provides equivalent fumigant action for extended periods of time. As such, they are advantageously employed as pesticidal agents for use in containers and cartons for stored food products such as, without limitation, flour, grain, wheat, barley, corn, pasta, cereal, pet food, and others.

The pesticidal compositions herein are so chemically inert that they are compatible with substantially any other constituents of stored products, and they may be used safely in the either the preparation of food products, or the application to the container, carton, or other packaging materials such as glue, after processing and/or packaging of the food products.

The term "carrier" as used herein means an inert or fluid material, which may be inorganic or organic and of synthetic or natural origin, with which the active compound is mixed or formulated to facilitate its application to the container or carton or other object to be treated, or its storage, transport and/or handling. In general, any of the materials customarily employed in formulating pesticides, herbicides, or fungicides, are suitable. The inventive pesticidal compositions of the present invention may be employed alone or in the form of mixtures with such solid and/or liquid dispersible carrier vehicles and/or other known compatible active agents such as other pesticides, or acaricides, nematocides, fungicides, bactericides, rodenticides, herbicides, fertilizers, growth-regulating agents, etc., if desired, or in the form of particular dosage preparations for specific application made therefrom, such as solutions, emulsions, suspensions, powders, pastes, and granules which are thus ready for use. The pesticidal compositions of the present invention can be formulated or mixed with, if desired,

conventional inert pesticide diluents or extenders of the type usable in conventional pesticide formulations or compositions, e.g. conventional pesticide dispersible carrier vehicles such as gases, solutions, emulsions, suspensions, emulsifiable concentrates, spray powders, pastes, soluble powders, dusting agents, granules, foams, pastes, tablets, aerosols, natural and synthetic materials impregnated with active compounds, microcapsules, coating compositions for use on seeds, and formulations used with burning equipment, such as fumigating cartridges, fumigating cans and fumigating coils, as well as ULV cold mist and warm mist formulations, etc.

Formulations containing the pesticidal compositions of the present invention may be prepared in any known manner, for instance by extending the pesticidal compositions with conventional pesticide dispersible liquid diluent carriers and/or dispersible solid carriers optionally with the use of carrier vehicle assistants, e.g. conventional pesticide surface-active agents, including emulsifying agents and/or dispersing agents, whereby, for example, in the case where water is used as diluent, organic solvents may be added as auxiliary solvents. Suitable liquid diluents or carriers include water, petroleum distillates, or other liquid carriers with or without surface active agents. The choice of dispersing and emulsifying agents and the amount employed is dictated by the nature of the composition and the ability of the agent to facilitate the dispersion of the pesticidal compositions of the present invention. Non-ionic, anionic, amphoteric, or cationic dispersing and emulsifying agents may be employed, for example, the condensation products of alkylene oxides with phenol and organic acids, alkyl aryl sulfonates, complex ether alcohols, quaternary ammonium compounds, and the like.

Liquid concentrates may be prepared by dissolving a composition of the present invention with a solvent and dispersing the pesticidal compositions of the present inventions in water with the aid of suitable surface active emulsifying and dispersing agents. Examples of conventional carrier vehicles for this purpose include, but are not limited to, aerosol propellants which are gaseous at normal temperatures and

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Surface-active agents, i.e., conventional carrier vehicle assistants, that may be employed with the present invention include, without limitation, emulsifying agents, such as non-ionic and/or anionic emulsifying agents (e.g. polyethylene oxide esters of fatty acids, polyethylene oxide ethers of fatty alcohols, alkyl sulfates, alkyl sulfonates, aryl sulfonates, albumin hydrolyzates, etc. and especially alkyl arylpolyglycol ethers, magnesium stearate, sodium oleate, etc.); and/or dispersing agents such as lignin, sulfite waste liquors, methyl cellulose, etc.

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the pesticide. These finely divided solids preferably have an average particle size of less than about 50 microns. A typical dust formulation useful for controlling insects contains 1 part of pesticidal composition and 99 parts of diatomaceous earth or vermiculite. Granules may comprise porous or nonporous particles. The granule particles are relatively large, a diameter of about 400-2500 microns typically. The particles are either impregnated or coated with the inventive pesticidal compositions from solution. Granules generally contain 0.05-15%, preferably 0.5-5%, active ingredient as the pesticidally-effective amount. Thus, the contemplated are formulations with solid carriers or diluents such as bentonite, fullers earth, ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, vermiculite, and ground synthetic minerals, such as highly-dispersed silicic acid, alumina and silicates, crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic materials such as sawdust, coconut shells, corn cobs and tobacco stalks. Adhesives, such as carboxymethyl cellulose, natural and synthetic polymers, (such as gum arabic, polyvinyl alcohol and polyvinyl acetate), and the like, may also be used in the formulations in the form of powders, granules or emulsifiable concentrations.

If desired, colorants such as inorganic pigments, for example, iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs or metal phthalocyanine dyestuffs, and trace elements, such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc may be used.

In commercial applications, the present invention encompasses carrier composition mixtures in which the pesticidal compositions are present in an amount substantially between about 0.01-95% by weight, and preferably 0.5-90% by weight, of the mixture, whereas carrier composition mixtures suitable for direct application or field application generally contemplate those in which the active compound is present in an amount substantially between about 0.0001-10%, preferably 0.01-1%, by weight of the

mixture. Thus, the present invention contemplates over-all formulations that comprise mixtures of a conventional dispersible carrier vehicle such as (1) a dispersible inert finely divided carrier solid, and/or (2) a dispersible carrier liquid such as an inert organic solvent and/or water, preferably including a surface-active effective amount of a carrier vehicle assistant, e.g. a surface-active agent, such as an emulsifying agent and/or a dispersing agent, and an amount of the active compound which is effective for the purpose in question and which is generally between about 0.0001-95%, and preferably 0.01-95%, by weight of the mixture.

The pesticidal compositions can also be used in accordance with so-called ultra-low-volume process, i.e. by applying such compounds or by applying a liquid composition containing the same, via very effective atomizing equipment, in finely divided form, e.g. average particle diameter of from 50-100 microns, or even less, i.e. mist form, for example by airplane crop spraying techniques. In this process it is possible to use highly concentrated liquid compositions with said liquid carrier vehicles containing from about 20 to 95% by weight of the pesticidal compositions or even the 100% active substances alone, e.g. about 20-100% by weight of the pesticidal compositions. The concentration in the liquid concentrate will usually vary from about 10 to 95 percent by weight and in the solid formulations from about 0.5 to 90 percent by weight.

Furthermore, the present invention encompasses methods for killing, combating or controlling pests, which comprises applying to at least one of correspondingly (a) such pests and (b) the corresponding habitat thereof, i.e. the locus to be protected, e.g. to a container or carton for stored food products, a correspondingly combative, a pesticidally effective amount, or toxic amount of the particular pesticidal compositions of the invention alone or together with a carrier as noted above. The instant formulations or compositions may be applied in any suitable usual manner, for instance by spraying, atomizing, vaporizing, scattering, dusting, watering, squirting, sprinkling, pouring, fumigating, and the like. The method for controlling beetles comprises applying the

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The pesticidal compositions and methods of the present invention are effective against a wide variety of beetles and it will be understood that the beetles exemplified and evaluated in the working Examples herein are representative of such a wider variety. For instance, the present invention can be used to control beetles that attack plants or warm-blooded animals, stored products and fabrics. Representative stored products that can be protected from pest attack by the present invention include, without limitation, grains, flour and flour products, tobacco and tobacco products, processed foods, cereals and the like. Representative fabrics that can be protected from pest attack by the invention are wool, cotton, silk, linen and the like.

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EXAMPLE 1

Pesticidal Effects of Plant Essential Oils As Fumigants Against Beetles

Various plant essential oils and blends thereof were tested for fumigant toxicity against maize weevil (*Sitophilus zeamais*) adults. The materials tested included 4-Blend (2-phenethyl alcohol, 2-phenethyl propionate, benzyl alcohol, and α -terpineol), benzyl alcohol, 2-phenethyl alcohol, 2-phenethyl propionate, trans-anethole, eugenol, 5-Blend (thymol, trans-anethole, eugenol, α -terpineol, and citronellal), α -terpineol, thymol, and V-3052 (eugenol, α -terpineol and cinnamic alcohol). To determine the fumigant action against stored grain insect pests, each of the test oils was fogged into a container in which adult maize weevils were present. The oils were applied at 200ug/cc, and maize weevil mortality was observed at 48 hours. The results are shown below.

48-hour mortality (%) at 200ug/cc

4-Blend	93%
Benzyl alcohol	93%
2-phenethyl alcohol	93%
2-phenethyl propionate	73%
Trans-anethole	53%
Eugenol	47%
5-Blend	40%
α -terpineol	20%
Thymol	13%
V-3052	7%

This data clearly demonstrates that certain plant essential oils and blends thereof may be used as a safe and effective alternative pesticide for control of beetles in containers and/or cartons for stored food products. The 4-Blend appears to exhibit synergistic effects when used as a fumigant when compared to the individual oils included in the blend.

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EXAMPLE 2

Pesticidal Effects of Plant Essential Oils As Fumigants Against Beetles

Various plant essential oils and blends thereof were screened for contact toxicity against maize weevil (*Sitophilus zeamais*) adults in corn, sawtoothed grain beetle (*Oryzaephilus surinamensis*) adults in oats, red flour beetle (*Tribolium castaneum*) adults in oats, and drugstore beetle (*Stegobium paniceum*) last-instar larvae in wheat. The materials tested included 4-Blend (2-phenethyl alcohol, 2-phenethyl propionate, benzyl alcohol, and α -terpineol), benzyl alcohol, ADL 1-19 (4-blend 10%, eugenol 1.7%, α -terpineol 1.7%, cinnamic alcohol 1.7%), ADL 1-22 (4-blend 10%, eugenol 2.5%, thymol 3%, cis-jasmone 0.6%), ADL 1-28 (2-phenethyl propionate 3.75%, thymol 3.0%, eugenol 2.5%, PD98059 0.03), and EcoPCO D (4-blend + eugenol). To determine the contact toxicity against stored insect pests, each of the test oils was formulated into a dust and then applied to a container in which the stored product and insect pests were present. The dusts were applied at different percentages relative to the stored products, and insect mortality was observed at 48 hours. The results are shown below.

<u>1. Maize Weevil Adults</u>	<u>% dust in corn</u>	<u>% mortality at 48 hrs.</u>
4-Blend	7	100
	3	100
	0.7	67
	0	0
Benzyl Alcohol	7	78
	3	44
	0.7	11
	0	0
ADL 1-19	7	100
	3	100
	0.7	78

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	0	0
ADL 1-22	7	100
	3	100
	0.7	100
	0	11
ADL 1-28	7	89
	3	89
	0.7	89
	0	0
EcoPCO D	7	100
	3	100
	0.7	89
	0	0

2. Sawtoothed Grain Adults	<u>% dust in oats</u>	<u>% mortality at 48 hrs.</u>
4-Blend	50	100
	25	100
	5	100
	0	0
Benzyl Alcohol	50	100
	25	100
	5	100
	0	0
ADL 1-19	50	100
	25	100
	5	80
	0	0

ADL 1-22	50	100
	25	100
	5	80
	0	0
ADL 1-28	50	100
	25	100
	5	50
	0	0
EcoPCO D	50	100
	25	100
	5	100
	0	0

3. Red Flour Beetle Adults % dust in oats % mortality at 48 hrs.

4-Blend	50	44
	25	22
	5	11
	0	0
Benzyl Alcohol	50	100
	25	89
	5	44
	0	0
ADL 1-19	50	56
	25	22
	5	0
	0	0
ADL 1-22	50	25
	25	40
	5	0

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	0	0
ADL 1-28	50	20
	25	11
	5	0
	0	0
EcoPCO D	50	20
	25	43
	5	33
	0	0
4. Drugstore Beetle Larvae	% dust in wheat	% mortality at 48 hrs.
4-Blend	10	13
	5	0
	1	11
	0	0
Benzyl Alcohol	10	100
	5	83
	1	0
	0	0
ADL 1-19	10	50
	5	67
	1	11
	0	0
ADL 1-22	10	63
	5	25
	1	67
	0	0
EcoPCO D	10	86
	5	33

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These data clearly demonstrate the pest specificity of these plant essential oils and how some blends are clearly synergistic in their action. These data also demonstrate the effective use rates for these plant essential oils and how best to deliver them. Further studies will be completed to evaluate the effective use of these compounds against different stages of the life cycle for these important stored product pests. The adult stage and last-instar larvae are the most difficult stages of life cycle to kill. If the plant essential oils are effective against any one stage, the life cycle will be successfully broken. Further studies are underway to evaluate the toxicity of these plant oils against the eggs and newly hatched larvae of these insect pests, which should be much more susceptible at this stage. Therefore, control of these two early stages can be accomplished at concentrations of insecticide lower than as shown in the data.

As can be seen from the above discussion, the pesticidal combinations of active compounds according to the present invention are markedly superior to known pesticidal agents/active compounds conventionally used for pest control in stored food product containers and/or cartons.

Although illustrative embodiments of the invention have been described in detail, it is to be understood that the present invention is not limited to those precise embodiments, and that various changes and modifications can be effected therein by one skilled in the art without departing from the scope and spirit of the invention as defined by the appended claims.